9th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 18–22, 2001

Spin coherence in semiconductor nanostructures

X. Marie†, A. Jbeli†, M. Paillard†, T. Amand† and J. M. Gérard‡
† Laboratoire de Physique de la Matiere Condensee, INSA-CNRS
135, Avenue de Rangueil, 31077 Toulouse cedex, France
‡ France Telecom, 196 avenue H. Ravera 92220 Bagneux, France

Abstract. We have studied the spin dynamics in self-organized InAs/GaAs quantum dots (QD) by time-resolved photoluminescence performed under strictly resonant excitation. We demonstrate that the carrier spins in these nanostructures are totally frozen on the exciton lifetime scale.

Introduction

A recent challenge in solid state physics is based on the manipulation of the electronic spin in order to build elementary quantum gates. The most important requirement for the operation of such future devices is a very long spin coherence time. Semiconductor QD appear as good candidates from this point of view. The discrete energy levels in artificial atoms like semiconductor quantum dots and the corresponding lack of energy dispersion lead indeed to a predicted modification of the spin relaxation dynamics compared to bulk or two-dimensional structures. Extensive experimental and theoretical works in the latter systems have identified the main carrier spin-flip mechanisms, which rely on exchange interaction, mixing between conduction and valence band states through spin-orbit coupling leading to motional narrowing effects in materials without inversion symmetry [1, 2]. In QD, all these processes have to be reconsidered since the wavevector k is no more a good quantum number [3, 4]. The absolute lack of energy states between QD energy levels is expected to suppress not only the elastic processes of spin relaxation but also the inelastic ones such as phonon scattering. The expected long spin relaxation time makes thus QD structures promising candidate for the implementation of spintronic and quantum information processing devices [5, 6]. Recent optical pumping experiments have indeed given good indications of a slowing down of the carrier spin relaxation processes in QD compared to bulk or quantum wells (QW) structures. The spin orientation in Wurtzite-type nanocrystals has been studied both in continuous wave (cw) and in time-resolved experiments [3, 7]. These investigations have revealed the QD size dependence of the spin polarization decay time. In naturally formed GaAs single QD, Gammon *et al.* deduced from cw photoluminescence (PL) experiments that the spin-flip scattering rate was lower than the radiative recombination rate of the ground state [8]. Similar experiments performed in self-organized InAs/GaAs QD lead to the same qualitative conclusion [9]. Recently, Gotoh *et al.* reported on an exciton spin relaxation time of about 900 ps in zero-dimensional InGaAs quantum disks, which is almost twice as long as the radiative recombination lifetime [10]. However most of these studies have been performed in non-resonant excitation conditions i.e. the QD polarization dynamics is studied when the spin-polarized carriers are photogenerated in the barrier. The observed spin polarization dynamics of the QD ground state is then the result of all the spin relaxation mechanisms which have occurred in the bulk barrier, in the QD excited state and finally in the QD ground state, including any spin flip scattering due to the energy relaxation process itself. In order to study the intrinsic spin dynamics, a time-resolved

strictly resonant excitation of the QD ground state is highly desirable [11]. Note that the free exciton radiative and the spin relaxation times in semiconductor QW have only been identified in such clear resonant excitation conditions [2, 12, 13]. We present here a detailed time-resolved investigation of the exciton optical alignment dynamics in self-organized InAs/GaAs QD performed under strictly resonant excitation. These experiments evidence a spin relaxation quenching at low temperature.

1. Samples and experimental setup

The investigated structure, grown by molecular beam epitaxy on a (001) GaAs substrate, consists of 5 InAs QD planes embedded in a GaAs λ planar cavity, inserted between two GaAs/AlAs Bragg mirror [14]. The QD layers are localized in the vicinity of the electromagnetic field antinodes into the microcavity which operates in the weak coupling regime. The QDs are obtained after a nominal deposition of 2.2 InAs monolayers and a 21 s growth interruption. These growth conditions lead to a QD density of $\sim 4 \times 10^{10}$ cm^{-2} per array, and to a QD emission centered around 1.15 eV with a Full Width at Half Maximum (FWHM) of ~60 meV. A variation of the cavity thickness along the radial direction of the wafer allows us to tune the cavity resonance by moving the laser beam across the sample surface. The microcavity is designed so that the cavity mode (FWHM ~3 meV) can be tuned in the energy range of the QD ground state emission. Inserting the QD in a microcavity brings two major improvements with respect to bare QD: (i) the filtering operated by the optical mode results in a reduction of the energetic inhomogeneity of the studied QD (ii) the narrowing of the radiation pattern emitted by the microcavity allows us to collect the QD emission efficiently in spite of the very small acceptance solid angle ($\sim 10^{-3}$ steradians) of the up-conversion detection set-up we have used [15].

The sample is excited by 1.5 ps pulses generated by an Optical Parametric Oscillator, synchronously pumped by a mode-locked Ti-doped sapphire laser. The time-resolved PL is then recorded by up-converting the luminescence signal in a $LiIO_3$ non-linear crystal with the picosecond pulses generated by the Ti:Sa laser [11, 15]. The time-resolution is limited by the laser pulse-width (~1.5 ps) and the spectral resolution is about 3 meV.

The linear and the circular polarization degrees of the luminescence are defined as $P_{\mathrm{Lin}} = (I^X - I^Y)/(I^X + I^Y)$ and $P_C = (I^+ - I^-)/(I^+ + I^-)$ respectively. Here I^X (I^Y) and I^+ (I^-) denote respectively the $X(Y)$ linearly polarized and the right (left) circularly polarized luminescence components (X and Y are chosen parallel to the [110] and [1$\bar{1}$0] sample directions). The experiments in section 2 and 3 have been performed at low excitation power (~7 W cm^{-2}) which corresponds to an average estimated density of photoexcited carriers less than one electron-hole pair per QD. In the following, the laser excitation energy is set to 1.137 eV; it coincides with both the cavity mode and a given QD family ground state energies. The detection energy is strictly the same as the excitation one [16].

Let us recall that in a (001)-grown type I quantum well, the relevant symmetry is D_{2d}. If the growth direction Oz is chosen as the quantization axis for the angular momentum, the conduction band is s-like, with two spin states $s_{e,z} = \pm 1/2$; the upper valence band is split into a heavy-hole band with the angular momentum projection $j_{hh,z} = \pm 3/2$ and a light-hole band with $j_{lh,z} = \pm 1/2$ at the center of the Brillouin zone. The heavy-hole exciton states can be described using the basis set $|J_z\rangle = |j_{hh,z}, s_{e,z}\rangle$, i.e.$|J_z = 1\rangle \equiv |3/2, -1/2\rangle$, $|J_z = -1\rangle \equiv |-3/2, 1/2\rangle$, $|J_z = 2\rangle \equiv |3/2, 1/2\rangle$, $|J_z = -2\rangle \equiv |-3/2, -1/2\rangle$ (J_z represents the total angular momentum projection of the exciton state on the quantization axis). This basis set is diagonal with respect to the exciton exchange interaction and the

twofold degenerate optically-active $J_z = |\pm 1\rangle$ states are split from the non-optically-active $J_z = |\pm 2\rangle$ states by the electron-hole exchange interaction energy [17].

In QD structures, the symmetry of the system is lowered and the exchange interaction is thus no longer isotropic [18]. The anisotropic exchange interaction splits the $|\pm 1\rangle$ radiative doublets into two eigenstates labeled $|X\rangle = (|1\rangle + |-1\rangle)/\sqrt{2}$ and $|Y\rangle = (|1\rangle - |-1\rangle)/(i\sqrt{2})$, linearly polarized along the [110] and [1$\bar{1}$0] directions respectively. Cw single dot spectroscopy experiments have clearly evidenced these two linearly polarized lines in self-organized InGaAs QD with an exchange splitting of $\hbar\omega \sim 150$ meV [9]. This anisotropic exchange splitting may originate from QD elongation [8] and/or interface optical anisotropy [18, 20].

2. Linear polarization dynamics at low temperature

Figure 1(a) displays the time-resolved PL intensity with polarization parallel (I^X) and perpendicular (I^Y) to the linearly polarized σ^X excitation laser ($T = 10$ K) (the initial peak on the I^X luminescence components intensity close to $t = 0$ corresponds to backscattered laser light from the sample surface). The corresponding linear polarization (P_{Lin}) kinetics is also plotted. The PL intensity decays with a characteristic time $\tau_{rad} \sim 800$ ps. After the pulsed excitation, the QD emission exhibits a strong linear polarization ($P_{\mathrm{Lin}} \sim 0.75$) which remains strictly constant within the experimental accuracy during the exciton emission (i.e. over ~2.5 ns). This behavior differs strongly from the exciton linear polarization dynamics in bulk or type I QW structures, characterized by a linear polarization decay time of a few tens of picoseconds [1, 15]. In the latter structures, it has been demonstrated that any scattering mechanism (phonon, exchange interaction, electron and/or hole single particle spin relaxation) results in the destruction of the coherent superposition between the $|1\rangle$ and $|-1\rangle$ components of the linear exciton [15]. The experimental observation of a QD exciton linear polarization which does not decay with time is the proof that neither the electron nor the hole spin relax on the exciton lifetime scale. It shows also that the exciton spin coherence between $|1\rangle$ and $|-1\rangle$ states is maintained during the whole exciton lifetime. From our observation, we can infer that the exciton spin relaxation time is longer than 20 ns, i.e. at least 25 times larger than the radiative lifetime.

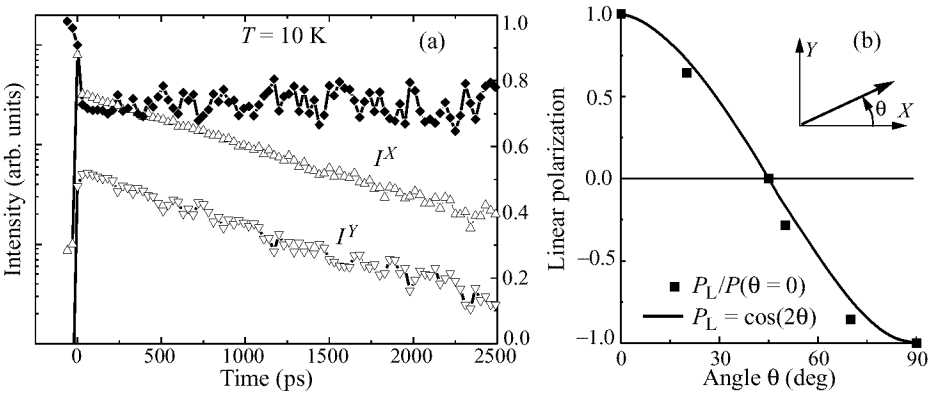

Fig. 1. (a) Time resolved PL intensity with polarization parallel I^X (Δ) and perpendicular I^Y (∇) to the linearly polarized (σ^X) excitation laser ($T = 10$ K); the time evolution of the corresponding linear polarization P_{Lin} is also displayed. (b) Dependence of the luminescence linear polarization on the angle of the excitation laser field with respect to the [110] direction (see text).

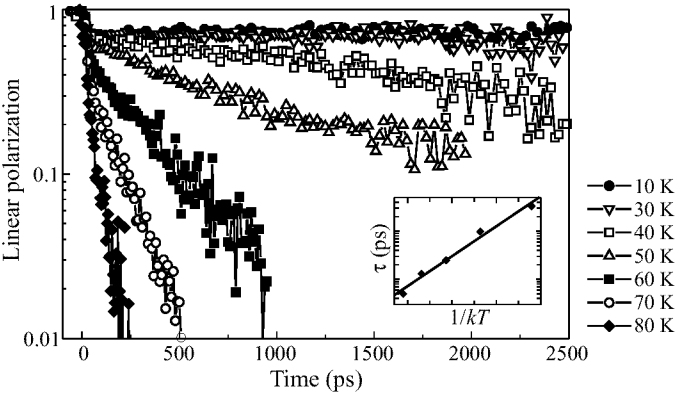

Fig. 2. Temperature dependence of the linear polarization dynamics (the excitation laser is σ^X polarized). Inset: P_{Lin} decay time as a function of $1/(K_B T)$.

Figure 2(b) displays the dependence of the luminescence linear polarization on the angle of the excitation laser field with respect to the [110] direction (see the inset).

The linearly polarized luminescence components are still detected along the [110] (X) and [1$\bar{1}$0] (Y) directions. As expected for exciton eigenstates polarized along the [110] and [1$\bar{1}$0] directions, the linear polarization follows a simple law with the angle θ given by:

$$P_{\mathrm{Lin}}(\theta) = \frac{I^X - I^Y}{I^X + I^Y} = \frac{\cos^2\theta - \sin^2\theta}{\cos^2\theta + \sin^2\theta} = \cos 2\theta.$$

A circularly polarized excitation should lead to the observation of P_{Lin} beats at the pulsation corresponding to the exchange splitting. After about 15 ps, the time required for the QD PL signal to overcome the backscattered laser light, we do not observe any linear or circular polarization and as a consequence any beat in this excitation configuration. This absence of polarization is interpreted as a consequence of the exchange splitting energy statistical fluctuations among the QD family selected by the cavity mode.

3. Temperature dependence of the linear polarization dynamics

Figure 2 presents the dependence of the exciton PL linear polarization dynamics as a function of the lattice temperature. A clear temporal decay of P_{Lin} is observed above 30 K: the linear polarization decay time drops from ~3500 ps at 40 K down to 50 ps at 80 K with an activation energy $E_a = 30 \pm 1$ meV (see the inset in Fig. 2). This very strong P_{Lin} temperature dependence can be due either to any electron, hole, exciton spin-flip scattering or any spin-conserving scattering processes which break the coherent superposition of the linearly photogenerated excitons $|X\rangle = (|1\rangle + |-1\rangle)/\sqrt{2}$ [21]. To the best of our knowledge there is no available theory on the exciton spin-flip and its temperature dependence in QD. However one can speculate that the depolarization mechanism is due to hole scattering to higher QD excited states since the measured activation energy is close to the energy splitting between the ground and first excited heavy-hole state and to the InAs LO phonon energy [22].

4. Excitation intensity dependence of the linear polarization dynamics

All the results presented up to now have been obtained at low excitation intensity (i.e. when the number of photo-generated electron-hole pairs is small compared to the num-

ber of QDs). Figure 3(a) displays the linear polarization dynamics under resonant linear polarization excitation for three excitation intensities. We note that the linear polarization never decays with time but the amplitude of this polarization decreases when the excitation intensity increases. This effect is due to the photo-generation of biexciton states. Under linearly polarized excitation, a quantum dot in which the ground state is already occupied by an exciton $|X\rangle = (|1\rangle + |-1\rangle)/\sqrt{2}$ can absorb another photon, which yields the photogeneration of a biexciton state labeled $(|XX + YY\rangle)/\sqrt{2}$. This biexciton state can emit σ^X or σ^Y photons with the same probability (the QDs occupied only by an exciton will still emit fully polarized σ^X luminescence). As a consequence, the amplitude of the measured linear polarization decays when the number of photogenerated biexciton states increases. We emphasize that the spectral resolution of our setup (~3 meV) does not allow to resolve separately the exciton and the biexciton emission lines.

This interpretation in terms of biexciton photogeneration is confirmed by the observed dependence of the total luminescence intensity on the excitation intensity displayed in Fig. 3(b). We observe that the luminescence intensity increases linearly with the excitation intensity at low excitation ($P < 10\ \mathrm{W\,cm^{-2}}$). In this regime when the number of photogenerated electron-hole pairs is small compared to the number of QDs, the photogeneration of biexciton states is negligible and the linear polarization does not depend on the excitation intensity. At higher excitation intensities ($P > 10\ \mathrm{W\,cm^{-2}}$), we note in Fig. 3(b) a clear saturation of the luminescence intensity and a simultaneous drop of the measured linear polarization value. The non-linear dependence of the luminescence intensity comes from

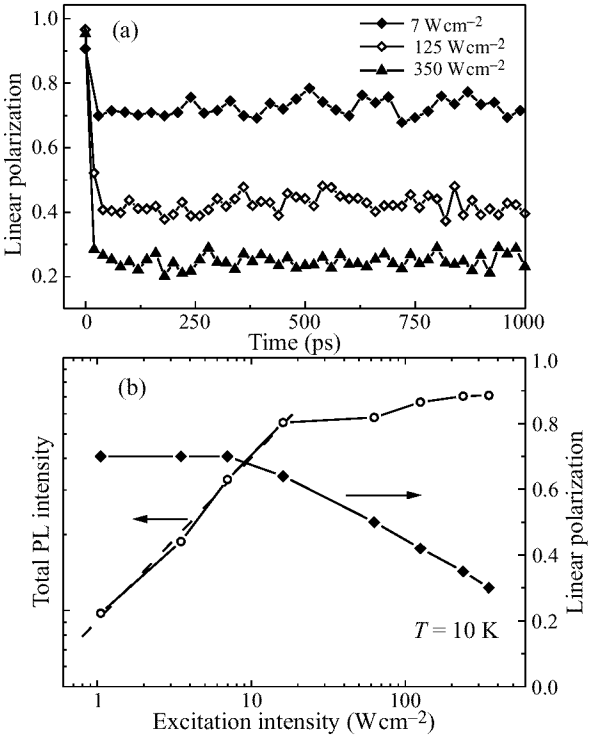

Fig. 3. (a) Linear polarization dynamics for three excitation laser powers ($T = 10$ K). (b) Total luminescence intensity and linear polarization as a function of the excitation power.

the saturation of the ground state absorption due to the photogeneration of biexciton states in most of the analysed QDs.

5. Conclusion

We have studied the linear PL polarization dynamics in self-organized QD under strictly resonant excitation. We never observe at low temperature any measurable temporal decay of the linear luminescence polarization regardless of the excitation intensity. This evidences a complete spin relaxation quenching in these zero-dimensional structures. This contrasts with the clear spin relaxation previously observed in non-resonant excitation conditions [10, 23]. The main difference between these two kinds of experiments relies on the higher energy carrier state occupation, including barrier, wetting layer and QD excited states which may induce the spin flip of the QD ground state by Coulomb exchange. The role of the carrier-carrier interaction in zero dimensional systems should be investigated theoretically to fully understand this difference.

The long spin coherence make these nanostructures an ideal candidate for the implementation of an elementary quantum gate in a solid state system.

Acknowledgments

We thank V. Thierry-Mieg for the sample growth. We are grateful to V. K. Kalevich, E. L. Ivchenko, K. V. Kavokin for fruitful discussions.

References

[1] *Optical Orientation*, eds F. Meier and B. Zakharchenya, *Modern Problems in Condensed Matter Sciences* Vol. 8 (North-Holland, Amsterdam, 1984).
[2] M. Z. Maialle et al., *Phys. Rev. B* **47**, 15 776 (1993).
[3] M. Chamaro et al., *J. of Lumin.* **70**, 222 (1996).
[4] A. V. Khaetskii et al., *Physica E* **6**, 470 (2000).
[5] R. Fiederling et al., *Nature* **402**, 787 (1999).
[6] A. Imamoglu et al, *Phys. Rev. Lett.* **83**, 4204 (1999).
[7] J. A. Gupta et al., *Phys. Rev. B* **59**, R10 421 (1999).
[8] D. Gammon et al., *Science* **273**, 87 (1996).
[9] M. Bayer et al., *Phys. Rev. Lett.* **82**, 1748 (1999).
[10] H. Gotoh et al. *Appl. Phys. Lett.* **72**, 1341 (1998).
[11] M. Paillard et al., *Appl. Phys. Lett.* **76**, 76 (2000).
[12] B. Deveaud et al., *Phys. Rev. Lett.* **67**, 2355 (1991).
[13] A. Vinattieri et al., *Phys. Rev. B* **50**, 10 868 (1994).
[14] J. M. Gerard et al., *Phys. Rev. Lett.* **81**, 1110 (1998).
[15] X. Marie et al., *Phys. Rev. Lett.* **79**, 3222 (1997).
[16] We did not notice any change in the presented effect when the excitation/detection energy was set to another value within the QD ground state energy distribution.
[17] T. Amand et al., *Phys. Rev. Lett.* **78**, 1355 (1997).
[18] E. L. Ivchenko, *Pure Appl. Chem.* **67**, 463 (1995); R. I. Dzhioev et al., *Phys. Solid State* **40**, 790 (1998).
[19] S. Hameau et al., *Phys. Rev. Lett.* **83**, 4152 (1999).
[20] R. I. Dzhioev et al., *Phys. Rev. B* **56**, 13 405 (1997); C. Gourdon et al., *Phys. Rev. B* **46**, 4644 (1992).
[21] M. Paillard et al., *Phys. Rev. Lett.* **86**, 1634 (2001).
[22] S. Sauvage et al., *Phys. Rev. B* **60**, 15 589 (1999).
[23] M. Paillard et al., *Phys. Stat. Sol.* (b) **221**, 71 (2000).